

Approximation of Reachable Set for Coherently Controlled Open Quantum Systems: Application to Quantum State Engineering

Jun Li,¹ Dawei Lu,² Zhihuang Luo,¹ Raymond Laflamme,^{2,3,4} Xinhua Peng,^{1,5,*} and Jiangfeng Du^{1,5,†}

¹*Hefei National Laboratory for Physical Sciences at Microscale and Department of Modern Physics, University of Science and Technology of China, Hefei, Anhui 230026, China*

²*Institute for Quantum Computing and Department of Physics and Astronomy, University of Waterloo, Waterloo, Ontario N2L 3G1, Canada*

³*Perimeter Institute for Theoretical Physics, Waterloo, Ontario N2L 2Y5, Canada*

⁴*Canadian Institute for Advanced Research, Toronto, Ontario M5G 1Z8, Canada*

⁵*Synergetic Innovation Center of Quantum Information & Quantum Physics, University of Science and Technology of China, Hefei, Anhui 230026, China*

Precisely characterizing and controlling realistic open quantum systems is one of the most challenging and exciting frontiers in quantum sciences and technologies. In this Letter, we present methods of approximately computing reachable sets for coherently controlled dissipative systems, which is very useful for assessing control performances. We apply this to a two-qubit nuclear magnetic resonance spin system and implement some tasks of quantum control in open systems at a near optimal performance in view of purity: e.g., increasing polarization and preparing pseudo-pure states. Our work shows interesting and promising applications of environment-assisted quantum dynamics.

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Recent years have seen immense advances in active and precise manipulation of a broad variety of quantum systems. The subject of quantum system control has been developed into a rapidly growing area [1] attracting substantial interests from the community of quantum information physicists. One of the fundamental tasks is to design reliable control techniques for systems that are exposed to a dissipative environment [2]. As dissipation tends to irreversibly affect the system dynamics, it is recognized as one dominant source for information loss and hence must be suppressed. Only recently was it realized that open system engineering may exhibit surprising advantages in some important aspects [3, 3, 5]. For example, it was shown that the purification efficiency of heat-bath algorithmic cooling protocol can surpass the closed system limit [3]. In other researches [5], there emerged great interests in environment-assisted entangled state engineering. Dissipative production of entangled steady states has already been realized in various experimental setups like trapped ions [6], superconducting circuit [7] and double quantum dot [8].

Although some ideas borrowed from classical control theory (e.g., time optimal control) have been successfully extended to construct methods for steering closed quantum systems [9], it turns out to be more challenging for open quantum systems. The major reason comes from the fact proved in [1] that for a finite dimensional Markovian quantum system, coherent means of control cannot fully compensate the irreversibility of the dynamics. In fact, to what extent can the system evolving tendency be changed depends upon not only the external operations but also the structure of the relaxation mechanisms. This certainly forms an obstacle in devising of general control

methodology. Previous research results have been able to characterize the reachable set on the states of a single qubit both qualitatively [1] and quantitatively [6, 12]. However, to generalize these results to higher dimensional systems is not easy [13].

Realising the lack of exact theory for the reachability problem, we propose to use approximation techniques. Basically the idea is to approximate the reachable set, usually in terms of simple geometric objects (e.g., convex polytopes [14], ellipsoids [15]), from both externally (over-approximation) and internally (under-approximation) [16]. Although various strategies have been put forward, computing reachable set in general remains a challenging task [14]. This is true especially for nonlinear systems, and quantum control models are indeed recognized as nonlinear [17]. In this Letter, we derive reasonable approximations of reachable set in coherently controlled Markovian quantum systems. To this end, we first study the upper bound of system purity function, thus giving an over-approximation that the system can not surpass; and then analyze the small time local controllability, which results in an under-approximation. Moreover, our ideas are implemented experimentally using techniques of nuclear magnetic resonance (NMR).

Problem setting—Consider a controlled n -qubit open system governed by the Lindblad equation [18, 19]

$$\dot{\rho} = -i[H_S + H_C(t), \rho] + \mathcal{R}\rho, \quad (1)$$

where H_S is the system Hamiltonian, $H_C(t)$ is the time-dependent external control Hamiltonian, and \mathcal{R} is the relaxation superoperator of Lindblad type. For simplicity we make two assumptions: (i) relaxation rates are comparatively slow so that arbitrary unitary operation can be

implemented before relaxation effects become important; (ii) system's free relaxation process leads to a strictly contractive channel \mathcal{E}_0 , namely the trace distance of any pair of different states is time decreasing. The latter assumption implies that, there is a unique relaxation-free state ρ_{eq} satisfying: $\mathcal{R}\rho_{eq} = 0$ [5]. Our assumptions are valid in many practical physical systems that are weakly interacted with a heat bath [19], e.g., atoms in a quantized radiation field and spin-lattice systems.

Now introduce the *vector of coherence representation* [2, 3]. Let $\mathcal{B} = \{B_k\}_{k=0}^{4^n-1} = \{I, X, Y, Z\}^{\otimes n}$, where I is the identity, and X, Y, Z are Pauli operators. It constitutes an orthonormal basis of the state space in that the orthonormal relation holds: $\text{Tr}(B_k B_j)/2^n = \delta_{kj}$ for $k, j = 0, \dots, 4^n - 1$. Consequently ρ can be expressed as: $\rho = I^{\otimes n}/2^n + \sum_{k=1}^{4^n-1} \mathbf{r}_k B_k$ ($\mathbf{r}_k = \text{Tr}(\rho B_k)/2^n$). The Lindblad Eq. (1) is then turned into a real $4^n - 1$ dimensional nonhomogeneous vector differential equation

$$\dot{\mathbf{r}} = \mathbf{H}\mathbf{r} - \mathbf{R}(\mathbf{r} - \mathbf{r}_{eq}), \quad (2)$$

in which \mathbf{H} , \mathbf{R} and \mathbf{r}_{eq} are $4^n - 1$ dimensional with their entries determined by $\mathbf{H}_{kj} = \text{Tr}(-iB_k[H_S + H_C, B_j])/2^n$, $\mathbf{R}_{kj} = \text{Tr}(-B_k \mathcal{R} B_j)/2^n$ and $\mathbf{r}_{eq,k} = \sum_j \mathbf{R}_{kj}^{-1} \text{Tr}(B_j \mathcal{R} I^{\otimes n})/4^n$ respectively. It can be verified that \mathbf{H} is antisymmetric and \mathbf{R} (relaxation matrix) is symmetric positive definite [22].

To further simplify the problem, we use the diagonalization procedure to project the system dynamics into the diagonal subspace spanned by $\{I, Z\}^{\otimes n}$ [6, 12]. Note that the vector of eigenvalues is essentially $2^n - 1$ dimensional. Let $\rho = U\Lambda U^\dagger$, where Λ is diagonal and U is a unitary operation in $SU(2^n)$. In the vector of coherence representation the diagonalization procedure can be written as $\mathbf{r} = \mathbf{U}\mathbf{x}$, where \mathbf{x} and \mathbf{U} are the representations of Λ and U with respect to basis \mathcal{B} respectively. Consequently, any evolution of the system can be projected into a continuous trajectory in the diagonal subspace. Substituting the diagonalization procedure into Eq. (2), we obtain a $2^n - 1$ dimensional dynamical equation [6]

$$\dot{\mathbf{x}} = -[\mathbf{U}^T \mathbf{R} \mathbf{U}]_{\mathbf{d}} \mathbf{x} + [\mathbf{U}^T \mathbf{R} \mathbf{r}_{eq}]_{\mathbf{d}}, \quad (3)$$

in which the notation $[\cdot]_{\mathbf{d}}$ denotes the diagonal subspace part of its argument. Provided that any unitary operation can be performed sufficiently fast compared with the relaxation timescale, we have that: (i) if a diagonal state can be reached, then any state on its unitary orbit can also be generated; (ii) according to Eq. (18), the system evolving direction at state \mathbf{x} can be adjusted to any element of the set $\{\dot{\mathbf{x}}_{\mathbf{U}} | \mathbf{U} \in SU(2^n)\}$. Thus it suffices to study the projected dynamics, and we can view $SU(2^n)$ as the admissible control set in place of $H_C(t)$. Let $\text{Reach}_{SU(2^n)}(\rho_{eq}, T)$ ($T \geq 0$) denote the reachable diagonal states from the equilibrium state ρ_{eq} under the control set $SU(2^n)$ during time $[0, T]$, the global reachable set is defined to be $\bigcup_{T \geq 0} \text{Reach}_{SU(2^n)}(\rho_{eq}, T)$.

Our goal is thus to construct both over-approximation and under-approximation of the reachable region of diagonal states. Clearly the problem here extends the concept of *universal bound on spin dynamics* [23], i.e., bounds on the regions of operators in Liouville space being interconvertible by unitary transformations, to the open system control regime. More precisely, let ρ and σ be two diagonal states, define the projection with respect to σ

$$\kappa_U = \text{Tr}(U\rho U^\dagger \cdot \sigma) / \text{Tr}(\sigma^2), \quad (4)$$

where $U \in SU(2^n)$, and

$$\kappa_{\mathcal{E}} = \text{Tr}(\mathcal{E}\rho \cdot \sigma) / \text{Tr}(\sigma^2), \quad (5)$$

where \mathcal{E} is the non-unitary channel given by Eq. (1) and satisfies $\mathcal{E}\rho = \kappa_{\mathcal{E}}\sigma$. Note that here in the latter case we don't allow the existence of unwanted components. When σ is the target operator, we can interpret κ_U and $\kappa_{\mathcal{E}}$ as the polarization transfer efficiency from ρ to σ . The universal bound gives an analytic expression bounding κ_U , which bears nice geometric meaning: the unitarily convertible region is bounded by a convex polytope whose vertexes are composed of all of the diagonal permutations of ρ . To move forward a step, we here study the extended problem of determining achievable regions for $\kappa_{\mathcal{E}}$.

Over-approximation— In over-approximating the reachable set, one identifies regions that the system can never reach. Our approach is to explore the dynamical behaviours of system purity function. Purity, quantifying the incoherent impacts from the environment, is particularly suited for studying how relaxation imposes restrictions on system evolution. For example, one basic result for *unital* systems, where ρ_{eq} is the maximally mixed state, states that the purity function must be monotonically decreasing with time regardless of the controls [24]. For the case of non-unital dynamics the situation is more complicated since purification can occur [1]. However, in practical situations, purification can not proceed unlimittedly, hence it is natural to seek for an upper bound.

Recall that purity is defined as $p = \text{Tr}\rho^2 = 1/2^n + 2^n \mathbf{r}^T \mathbf{r}$, thus its first time derivative is given by

$$\dot{p} = -2^{n+1} \mathbf{r}^T \mathbf{R}(\mathbf{r} - \mathbf{r}_{eq}). \quad (6)$$

The set of states satisfying $\dot{p} = 0$ determines an ellipsoid in \mathbb{R}^{4^n-1} , which depends only upon \mathbf{R} and the equilibrium state. From positive definiteness of \mathbf{R} we know that for any state \mathbf{r} outside of the ellipsoid there must be $\dot{p}(\mathbf{r}) < 0$. Let S denote the smallest sphere enclosing the ellipsoid, it is obvious that: (i) the state \mathbf{r}_{eq} is located inside S and (ii) the evolution direction of any state on S is towards the inner side of S . Thus starting at \mathbf{r}_{eq} , the system can not be driven outside S by coherent means. One can then envisage a simple method to get an upper bound of p by solving the following optimization problem

$$\begin{cases} \max & p(\mathbf{r}) = 1/2^n + 2^n \mathbf{r}^T \mathbf{r}, \\ \text{s.t.} & \dot{p}(\mathbf{r}) = -2^{n+1} \mathbf{r}^T \mathbf{R}(\mathbf{r} - \mathbf{r}_{eq}) = 0. \end{cases}$$

This problem can be seen as an instance of *quadratic programming over an ellipsoid constraint*, which is easy in the sense of computational complexity and can be solved with well-developed algorithms [25].

Under-approximation—Under-approximation involves some simplifications of the problem, which we state as such: (i) we restrict our considerations to the discrete set of controls $\mathcal{Q} \subset SU(2^n)$, where \mathcal{Q} is the collection of $2^n!$ permutation operations on diagonal elements of the density matrix; (ii) we will find the small-time local controllable (STLC) set of states rather than analyzing global controllability. The system is said to be *small-time local controllable* at point \mathbf{x} if \mathbf{x} belongs to the interior of the reachable set $\text{Reach}_{SU(2^n)}(\mathbf{x}, T)$ for all $T > 0$. In other words, for STLC at a point we need to be able to generate small motions in any direction of the full space \mathbb{R}^{2^n-1} at that point. We now use $\Omega_{\mathcal{Q}}$ to denote the STLC set under the discrete control set \mathcal{Q} .

The problem of analytically constructing $\Omega_{\mathcal{Q}}$ was studied in full length in Ref. [13], with the conclusion that $\Omega_{\mathcal{Q}}$ is open, compact and connected, and its boundary is composed of a number of hypersurfaces. Knowing about the connectedness of $\Omega_{\mathcal{Q}}$, along with the definition of STLC, we can take $\Omega_{\mathcal{Q}}$ as an under-approximation

$$\Omega_{\mathcal{Q}} \subset \bigcup_{T \geq 0} \text{Reach}_{SU(2^n)}(\rho_{eq}, T). \quad (7)$$

An algorithmic procedure of calculating $\Omega_{\mathcal{Q}}$ is presented in Supplemental Material [22].

Applications on two-qubit system.— We use the ^{13}C -labeled chloroform dissolved in d_6 -acetone as a two-qubit system to test the applicability of our reachability analysis. Our experiments were carried out on a Bruker Avance III 400 MHz ($B_0 = 9.4$ T) spectrometer at room temperature. The natural Hamiltonian at the basis $\mathcal{B}_2 = \{I, X, Y, Z\}^{\otimes 2}$ reads: $H_S = \pi(-\gamma_C B_0 Z I - \gamma_H B_0 I Z + J/2 Z Z)$, where γ_C and γ_H are the gyromagnetic ratios of nucleus ^{13}C and ^1H respectively, and $J = 214.5\text{Hz}$ is the scalar coupling constant. The equilibrium state is of the form: $\rho_{eq} \approx II/4 + \epsilon_C Z I + \epsilon_H I Z$ with $\epsilon_C \approx \epsilon_H/4 \equiv \epsilon \sim 10^{-5}$. In the double rotating frame, we measured all the effective relaxation rates and thus obtain the system relaxation matrix \mathbf{R} [22].

In order to visualize the system evolution, we project the 15-dimensional relaxation dynamics into a 3-dimensional differential equation according to Eq. (18). For simplicity, we focus on the region $0 \leq \mathbf{x}_3 \leq \mathbf{x}_1 \leq \mathbf{x}_2$ in which our control experiments are performed. In the region, we derived the sphere S representing the upper bound of system purity and the surface E representing the boundary for the system STLC set under the discrete control set \mathcal{Q} based on the measured relaxation matrix (see Fig. 1(a)). The boundary for the exact reachable set should thus be in between S and E . We also plotted the faces of the polygon, denoted by P , representing the universal bound on spin dynamics under unitary

control. It can be seen that, in this system, the exact reachable set exceeds the unitary universal bound in almost every direction, clearly demonstrating the possibility of larger reachable region of states when relaxation is present. Now we will study how the obtained results help us gain insights into the open system control methods.

Our first concern is the intersection between S and the \mathbf{x}_2 axis: $(0, 4.27\epsilon, 0)$. We can use the nuclear Overhauser effect (NOE) to approach this state. It is well-known that [34], for a heteronuclear two-spin system, applying a field at the resonance frequency of one spin for a sufficiently long time, will saturate its polarization and at the meantime affect or even enhance the magnetization of the other spin. In the experiment, an irradiation with 1000Hz of magnitude and 10s of duration is applied to the carbon channel, which drives the system into a steady state measured as $\mathbf{x}_{ss} \approx (0, 4.25\epsilon, 0)$. The ^1H polarization is enhanced and is fairly close to the upper bound (point A_2 in Fig. 1(a)), in contrast to the unitary bound.

Our purity bound analysis thus leads to a new view of the NOE experiment. The polarization transfer efficiency in NOE experiment is nearly optimal, which shows the advantages of environment-assisted quantum control. Moreover, it extends the results of algorithmic cooling schemes. According to the purification limits derived in [3], it is impossible to cool the proton in our system through the “compression and refresh” iterative procedure. This is due to the different underlying relaxation model assumed. In heat-bath algorithmic cooling scheme, it is considered that each qubit is undergoing their respective T_1 and T_2 processes. But in NOE, cross-relaxation mechanisms are essential for the ^1H purification [34]. Thus NOE provides clear evidence of approaching even larger purification efficiency if more general relaxation mechanisms are taken into account.

Next we turn to the application of open system coherent control to state engineering in NMR quantum computation. We consider creating pseudopure state (PPS) [27–29] from the equilibrium state, which is an often used initialization step for subsequent computation. The task can not be done merely with unitary operations. Previous methods of PPS preparation involve different ways of realizing non-unitary operations [30] such as exertion of gradient fields. Here, we put forward a new approach: to let the inherent system relaxation effects take the role of non-unitary resources and design a periodic sequence so that PPS is the fixed point of the dynamics. Although the current experiment is performed on two-qubit system as an example, the idea can apply to general cases.

For chloroform, PPS takes the form: $\rho_{pps} = II/4 + \eta/4(ZI + IZ + ZZ)$, in which η is the *effective purity*. The feature that its three coefficients are equal to each other specifies the *PPS direction*, namely $\mathbf{x}_1 = \mathbf{x}_2 = \mathbf{x}_3$. Therefore, it is straightforward to conceive a simple “coefficient-averaging process”. The averaging process is governed by $[\tau - \mathbf{V}]_m$, where m is the iteration number,

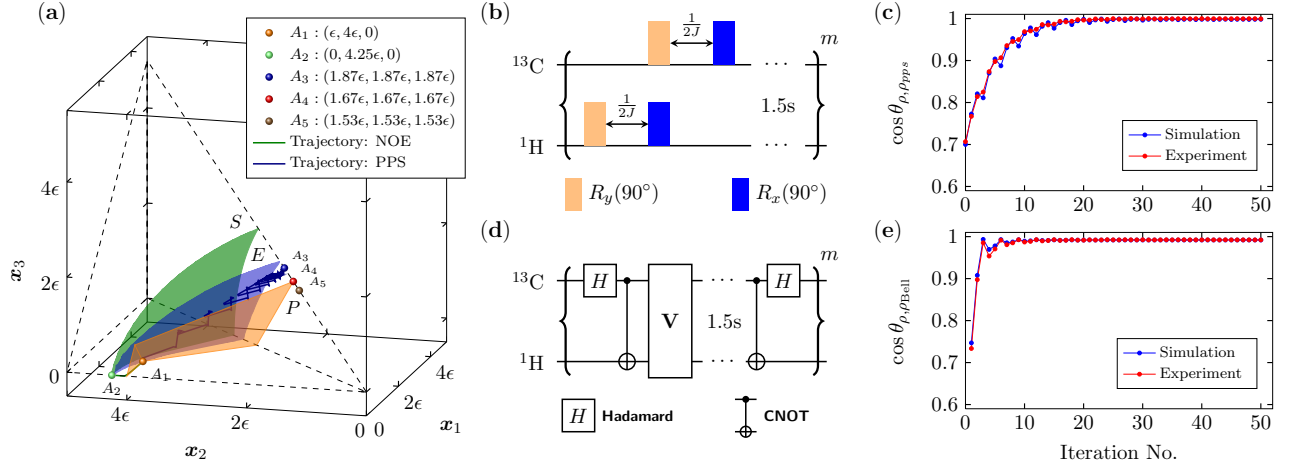


FIG. 1: (a) Illustrations of the results (in the region $0 \leq x_3 \leq x_1 \leq x_2$) on our chloroform system, including: (i) sphere (green) $S : \mathbf{x}^T \mathbf{x} = 18.06\epsilon^2$; (ii) surface (blue) E : boundary for STLC region under control set \mathcal{Q} ; (iii) faces (orange) P representing the unitary universal bound; (iv) projected trajectories (simulation) for PPS preparation and NOE experiments. (b)-(c) The resulting PPS ρ_{pps} , and (d)-(e) pseudo-Bell state ρ_{Bell} under periodic controls, along with numerical simulations with the Lindblad equation Eq. (2). Here $\theta_{\rho, \rho_{pps}}$ and $\theta_{\rho, \rho_{Bell}}$ denotes the angle between the direction of ρ and that of the desired states. As long as the excitation is on, the system will be preserved periodically at the desired state direction.

τ represents a period of free relaxation, and \mathbf{V} is a cyclic permutation of the coordinates of \mathbf{x} . Let \mathcal{E}_τ and $\mathcal{E}_\mathbf{V}$ denote the dynamic map associated with the τ relaxation evolution and \mathbf{V} operation respectively. Provided that τ is small enough, namely \mathcal{E}_τ is close to the identity, then the fixed point of $\mathcal{E}_\mathbf{V} \circ \mathcal{E}_\tau$ is close to that of $\mathcal{E}_\mathbf{V}$. In the experiments, we chose $\mathbf{V} : (x_1, x_2, x_3)^T \rightarrow (x_2, x_3, x_1)^T$, which is implemented through a simple sequence shown in Fig. 1(b). It was found that for a wide range of τ (less than ~ 2 s) the system was able to be driven to some states close to the PPS direction. Within tolerable range of error, we set $\tau = 1.5$ s, giving the maximal effective purity ($\eta \approx 7.48\epsilon$, point A_3 in Fig. 1(a)) of PPS obtained on trials. This can be compared to conventional spatial averaging method where $\eta \approx 6.12\epsilon$ (point A_5 in Fig. 1(a)) [31] and line-selective pulse approach where unitary bound can be achieved $\eta \approx 6.67\epsilon$ (point A_4 in Fig. 1(a)) [32]. Fig. 1(a) also reveals a gap between the point of maximally reachable η (in between over-approximation and under-approximation) and the obtained PPS. The gap can be attributed to several reasons: (i) the imprecision in the experimental estimation of the relaxation matrix \mathbf{R} ; (ii) the assumption of ignoring relaxation effects during the operation \mathbf{V} is not perfectly satisfied in practice; (iii) the over-approximation may be not sufficiently tight and there is also the possibility that better preparation method exists.

Our periodic control method applies to any state that is unitarily equivalent to a PPS, e.g., a pseudo-Bell state $\rho_{Bell} = (1-\eta)/4II + \eta/2(|00\rangle + |11\rangle) \otimes (|00\rangle + |11\rangle)$. The trick is that, we just modify the PPS preparation periodic sequence to be $[\mathbf{W} - \tau - \mathbf{V} - \mathbf{W}^T]_m$ (Fig. 1(d)), so that ρ_{Bell} now becomes the fixed point of the sequence.

Here \mathbf{W} transforms ρ_{pps} to ρ_{Bell} and can be implemented through a Hadamard gate and a CNOT gate. The experimental results shown in Fig. 1(e) demonstrates that the proposed control method can be implemented and gives results in excellent agreement with predictions.

The theory presented here helps assessing open system control schemes where purity is an important metric. In addition, our approximation can guide the development of numerical pulse searching algorithms. Our work can be improved by increasing the efficiency of computing the approximation, with the aid of advanced algorithmic techniques from computational geometry. We further studied in detail the NOE effect and state engineering experiments in the open system framework, and showed that relaxation effects are useful for implementing some nontrivial non-unitary control tasks. The lack of full controllability in certain important control regimes [33, 34] usually calls for a bound analysis for system reachable states. Our present study can thus be regarded as a part of explorations in this direction. Future work will concentrate on incorporating our work here with other open system control models [1], such as reservoir engineering in which incoherent resources are introduced to enhance the capability of controlling quantum systems.

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* Electronic address: xhpeng@ustc.edu.cn

† Electronic address: djf@ustc.edu.cn

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Lindblad Equation in Vector of Coherence Representation

Here we rewrite Lindblad equation in the vector of coherence representation, basically following the discussions in [1–3]. Introducing the orthonormal basis $\mathcal{B} = \{B_k\}_{k=0}^{4^n-1} = \{I, X, Y, Z\}^{\otimes n}$, then there is

$$\rho = I^{\otimes n}/2^n + \sum_{k=1}^{4^n-1} \mathbf{r}_k B_k, \quad \mathbf{r}_k = \text{Tr}(\rho B_k)/2^n. \quad (8)$$

Substituting the above expression into the Lindblad Eq. (1) yields

$$\begin{aligned} \dot{\mathbf{r}}_k &= \text{Tr}(B_k(-i[H, \rho] + \mathcal{R}\rho))/2^n \\ &= \text{Tr}\left(-iB_k\left[H, \sum_{j=1}^{4^n-1} \mathbf{r}_j B_j\right]\right)/2^n + \text{Tr}\left(B_k\mathcal{R}\left(\frac{I^{\otimes n}}{2^n} + \sum_{j=1}^{4^n-1} \mathbf{r}_j B_j\right)\right)/2^n \\ &= \sum_{j=1}^{4^n-1} \text{Tr}(-iB_k[H, B_j])/2^n \mathbf{r}_j + \sum_{j=1}^{4^n-1} \text{Tr}(B_k\mathcal{R}B_j)/2^n \mathbf{r}_j + \text{Tr}\left(B_k\mathcal{R}\left(\frac{I^{\otimes n}}{2^n}\right)\right)/2^n. \end{aligned} \quad (9)$$

Define

$$\mathbf{H}: \quad \mathbf{H}_{kj} = \text{Tr}(-iB_k[H, B_j])/2^n, \quad (10)$$

$$\mathbf{R}: \quad \mathbf{R}_{kj} = \text{Tr}(-B_k\mathcal{R}B_j)/2^n, \quad (11)$$

$$\mathbf{v}: \quad \mathbf{v}_k = \text{Tr}(B_k\mathcal{R}I^{\otimes n})/4^n. \quad (12)$$

Then we get

$$\dot{\mathbf{r}} = \mathbf{H}\mathbf{r} - \mathbf{R}\mathbf{r} + \mathbf{v}. \quad (13)$$

Proposition 1. \mathbf{R} is real, symmetric. If the pure relaxation process of the system is strictly contractive, then \mathbf{R} is positive definite.

Proof. In general \mathbf{R} is self-adjoint, the proof of which can be found in [2, 4]. Provided that ρ is decomposed with respect to the generalized Pauli basis $\{B_k\}_{k=0}^{4^n-1}$, then as $\mathcal{R}B_j$ is self-adjoint, $\text{Tr}(-B_k\mathcal{R}B_j)$ should be real for all $k, j = 1, \dots, 4^n - 1$. So \mathbf{R} is real and hence symmetric.

If the pure relaxation process leads to a strictly contractive channel, then there exists a unique fixed point, which we denote by \mathbf{r}_0 ([5], page 408). It is the unique solution to the equation $(\mathbf{H}_S - \mathbf{R})\mathbf{r}_0 + \mathbf{v} = 0$. Thus $\mathbf{H}_S - \mathbf{R}$ should be of full rank such that $\mathbf{r}_0 = (-\mathbf{H}_S + \mathbf{R})^{-1}\mathbf{v}$. The pure relaxation dynamics is then described by

$$\dot{\mathbf{r}} = (\mathbf{H}_S - \mathbf{R})(\mathbf{r} - \mathbf{r}_0).$$

Furthermore, the property of strictly contractiveness implies that, for any state $\mathbf{r}(t)$ other than \mathbf{r}_0 , the time derivative of the trace distance of which to \mathbf{r}_0 should satisfy

$$\dot{D}(\mathbf{r}(t), \mathbf{r}_0) = \frac{1}{2} \frac{d|\mathbf{r} - \mathbf{r}_0|}{dt} = -\frac{(\mathbf{r} - \mathbf{r}_0)^T \mathbf{R}(\mathbf{r} - \mathbf{r}_0)}{4|\mathbf{r} - \mathbf{r}_0|} < 0.$$

This condition can be fulfilled only if \mathbf{R} is symmetric positive definite. \square

Since we have assumed at the beginning that our considered system under pure relaxation process is strictly contractive, \mathbf{R} should be of full rank by the above proposition, thus if we define

$$\mathbf{r}_{eq} = \mathbf{R}^{-1}\mathbf{v}, \quad (14)$$

then accordingly we will have

$$\dot{\mathbf{r}} = \mathbf{H}\mathbf{r} - \mathbf{R}(\mathbf{r} - \mathbf{r}_{eq}). \quad (15)$$

Derivation of Eq. (4)

The derivation of the projected dynamic equation can actually be found in [6], which we copy as follows.

At each instant of time, we can diagonalize the density matrix $\rho(t) = U(t)\Lambda(t)U^\dagger(t)$ by a unitary matrix $U(t)$. Substitute it into the Lindblad Eq. (1), we get

$$\begin{aligned}\dot{\Lambda}(t) &= \dot{U}^\dagger(t)\rho(t)U(t) + U^\dagger(t)\dot{\rho}(t)U(t) + U^\dagger(t)\rho(t)\dot{U}(t) \\ &= iU^\dagger(t)H'(t)\rho(t)U(t) + U^\dagger(t)\{-i[H(t), \rho(t)] + \mathcal{R}\rho(t)\}U(t) - U^\dagger(t)\rho(t)iH'(t)U(t) \\ &= -iU^\dagger(t)[H(t) - H'(t), \rho(t)]U(t) + U^\dagger(t)\mathcal{R}(\rho(t))U(t), \\ &= -iU^\dagger(t)[H(t) - H'(t), U(t)\Lambda(t)U^\dagger(t)]U(t) + U^\dagger(t)\mathcal{R}(U(t)\Lambda(t)U^\dagger(t))U(t),\end{aligned}$$

where we have defined $\dot{U}(t) = -iH'(t)U(t)$, and $H'(t)$ must be Hermitian by the fact that $d(U(t)U^\dagger(t))/dt = 0$.

Note that the left side of the above equation is a diagonal matrix, so for the right side we only need to keep the diagonal part. Moreover, the first term on the right side is a commutation of two Hermitian matrices, and since $\Lambda(t)$ is diagonal, so the diagonal part of this commutation must be zero. Therefore, the above equation reduces to

$$\dot{\Lambda}(t) = \text{diag}(U^\dagger(t)\mathcal{R}(U(t)\Lambda(t)U^\dagger(t))U(t)). \quad (16)$$

Now we go to the vector representation.

$$\begin{array}{ccc} \rho & \xleftarrow{U(t)} & \Lambda \\ \downarrow & & \downarrow \\ \mathbf{r} & \xleftarrow{\mathbf{U}(t)} & \mathbf{x} \end{array}$$

Substitute $\mathbf{r} = \mathbf{U}(t)\mathbf{x}$ into Eq. (15), there is

$$\begin{aligned}\dot{\mathbf{x}} &= \mathbf{U}^T \mathbf{H} \mathbf{U} \mathbf{x} - \mathbf{U}^T \mathbf{R} \mathbf{U} \mathbf{x} + \mathbf{U}^T \mathbf{R} \mathbf{r}_{eq} \\ &= [\mathbf{U}^T \mathbf{H} \mathbf{U} \mathbf{x} - \mathbf{U}^T \mathbf{R} \mathbf{U} \mathbf{x} + \mathbf{U}^T \mathbf{R} \mathbf{r}_{eq}]_{\mathbf{d}}.\end{aligned}$$

As has been just demonstrated, the first term of the above equation should vanish, thus

$$\dot{\mathbf{x}} = -[\mathbf{U}^T \mathbf{R} \mathbf{U}]_{\mathbf{d}} \mathbf{x} + [\mathbf{U}^T \mathbf{R} \mathbf{r}_{eq}]_{\mathbf{d}}, \quad (17)$$

Under-approximation of the Reachable Set

The projected dynamics in the diagonal subspace goes

$$\dot{\mathbf{x}} = -[\mathbf{U}^T \mathbf{R} \mathbf{U}]_{\mathbf{d}} \mathbf{x} + [\mathbf{U}^T \mathbf{R} \mathbf{r}_{eq}]_{\mathbf{d}}, \quad (18)$$

in which \mathbf{U} runs over all elements of the group $SU(2^n)$. For each $\mathbf{U} \in SU(2^n)$, there corresponds to an evolving direction. Since we assume that any unitary operation can be performed very fast compared with the relaxation timescale, the system evolving direction at state \mathbf{x} can be adjusted to any element of the set

$$\{\dot{\mathbf{x}}_{\mathbf{U}} = -[\mathbf{U}^T \mathbf{R} \mathbf{U}]_{\mathbf{d}} \mathbf{x} + [\mathbf{U}^T \mathbf{R} \mathbf{r}_{eq}]_{\mathbf{d}} | \mathbf{U} \in SU(2^n)\}. \quad (19)$$

Now we want to under-approximate the system reachable set. To this end, we study the simplified reachability problem: (i) instead of considering the whole control set $SU(2^n)$, we restrict our attention to the discrete set of controls \mathcal{Q} ; (ii) we will find the small-time local controllable set of states rather than analyzing global controllability.

For system (18) under the discrete control set \mathcal{Q} , we denote the set of admissible evolving directions at an arbitrary state \mathbf{x} as

$$\{\dot{\mathbf{x}}_{\mathbf{Q}_k} = -[\mathbf{Q}_k^T \mathbf{R} \mathbf{Q}_k]_{\mathbf{d}} \mathbf{x} + [\mathbf{Q}_k^T \mathbf{R} \mathbf{r}_{eq}]_{\mathbf{d}} | \mathbf{Q}_k \in \mathcal{Q}\}. \quad (20)$$

Let $\text{cone}(\{\dot{\mathbf{x}}_{\mathbf{Q}_k}\})$ be the convex cone generated by the vector fields $\{\dot{\mathbf{x}}_{\mathbf{Q}_k}\}$ through conical combination:

$$\text{cone}(\{\dot{\mathbf{x}}_{\mathbf{Q}_k}\}) = \left\{ \sum_{k=1}^{2^n!} c_k \dot{\mathbf{x}}_{\mathbf{Q}_k} \mid c_k \geq 0, \mathbf{Q}_k \in \mathcal{Q} \right\}. \quad (21)$$

Then ([7], page 56),

Proposition 2. *Given the discrete set of admissible vector fields $\{\dot{\mathbf{x}}_{\mathbf{Q}_k}\}$, one can and only can generate motions in the convex cone $\text{cone}(\{\dot{\mathbf{x}}_{\mathbf{Q}_k}\})$.*

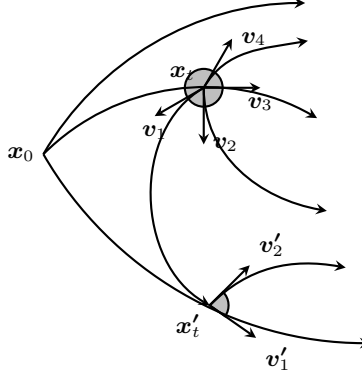


FIG. 2: Illustration of STLC property. It can be seen that the cone generated by the velocity vectors at \mathbf{x}_t is the full space, while this is not true for \mathbf{x}'_t .

Let $\text{Reach}_{\mathcal{Q}}(\mathbf{x}, T)$ ($T > 0$) denote the reachable set from state \mathbf{x} under control \mathcal{Q} during time $[0, T]$. We say system (18) is *small-time local controllable (STLC)* at point \mathbf{x} if \mathbf{x} belongs to the interior of the reachable set $\text{Reach}_{\mathcal{Q}}(\mathbf{x}, T)$ for all $T > 0$. In other words, for STLC at a point we need to be able to generate small motions in any direction of the full space \mathbb{R}^{2^n-1} at that point. So one has that, system (18) is STLC at point \mathbf{x} iff $\text{cone}(\{\dot{\mathbf{x}}_{\mathbf{Q}_k}\}) = \mathbb{R}^{2^n-1}$. We denote the system STLC set under the discrete control set \mathcal{Q} by $\Omega_{\mathcal{Q}}$.

Constructing the STLC Set

P. Rooney analytically constructed the STLC set under the control set \mathcal{Q} in Ref. [7]. It turns out that $\Omega_{\mathcal{Q}}$ is open, compact and connected, and its boundary is composed of a number of surfaces. We here just copy the core result obtained by P. Rooney:

Theorem ([7], page 94). *For every subset $\sigma \subset \{1, 2, \dots, 2^n!\}$ with $2^n - 1$ elements, construct the hypersurface $\mathbf{x}_{\sigma} = (\sum_{k \in \sigma} \mu_k [\mathbf{Q}_k^T \mathbf{R} \mathbf{Q}_k]_{\mathbf{d}})^{-1} (\sum_{k \in \sigma} \mu_k [\mathbf{Q}_k^T \mathbf{R} \mathbf{r}_{eq}]_{\mathbf{d}})$, where $\mu_k \geq 0$ and $\sum_{k \in \sigma} \mu_k = 1$. Denote $\bigcup_{\sigma} \mathbf{x}_{\sigma}$ as the union of all such hypersurfaces, which is a closed hypersurface. Then $\Omega_{\mathcal{Q}}$ is an open set whose closure is equal to the closure of $\bigcup_{\sigma} \mathbf{x}_{\sigma}$.*

Algorithm for Testing STLC

In practice, it is extremely difficult to compute $\Omega_{\mathcal{Q}}$. Just take two-qubit system as an example, the number of surfaces to be computed is $C_{24}^4 = 10626$, and an extra effort of making the union of these surfaces has to be made. Here in order to compute the boundary of $\Omega_{\mathcal{Q}}$ in the case of a two-qubit system, we choose an alternative approach that can reduce practical computing efforts to a large extent. Basically, we sample the state space with a discrete set of points, and choose those STLC points among them through a STLC testing algorithm.

To determine whether a vector is in a convex cone, belongs to the class of point-in-polygon problems, and there exist a number of algorithms to solve them in the realm of computational geometry. Here we present a testing algorithm based on the so called *fundamental theorem of linear inequalities* (this classic theorem is due to Farkas, Minkowski, Carathéodory, Weyl, etc.) [8].

Theorem (Fundamental Theorem of Linear Inequalities). *Let $\{\mathbf{v}_k\}$ and \mathbf{u} be vectors in \mathbb{R}^m , and suppose $\text{span}(\{\mathbf{v}_k\}) = \mathbb{R}^m$. Then exactly one of the two statements is true: (i) $\mathbf{u} \in \text{cone}(\{\mathbf{v}_k\})$; (ii) there exists a hyperplane $\{\mathbf{x} | \mathbf{n} \cdot \mathbf{x} = 0\}$, containing $m - 1$ linearly independent vectors from $\{\mathbf{v}_k\}$, such that $\mathbf{n} \cdot \mathbf{u} > 0$ and $\mathbf{n} \cdot \mathbf{v}_k \leq 0$ for all k , which means that there is a hyperplane spanned by $m - 1$ vectors from $\{\mathbf{v}_k\}$ (with \mathbf{n} being its normal vector) separating $\text{cone}(\{\mathbf{v}_k\})$ and \mathbf{u} .*

The theorem provides a criterion to test whether a given set of vector fields is STLC.

Corollary 1. *Let $\{\mathbf{v}_k\}$ be vectors in \mathbb{R}^m , and $\text{span}(\{\mathbf{v}_k\}) = \mathbb{R}^m$. Then $\text{cone}(\{\mathbf{v}_k\}) = \mathbb{R}^m$, iff for any hyperplane spanned by $m - 1$ linearly independent vectors from $\{\mathbf{v}_k\}$, its normal vector \mathbf{n} satisfies that $\{\mathbf{n} \cdot \mathbf{v}_k\}$ is not all nonpositive and not all nonnegative.*

Proof. Positive direction. It is evident from the “fundamental theorem of linear inequalities”.

Inverse direction. If there exists a nonzero vector \mathbf{n} such that $\{\mathbf{n} \cdot \mathbf{v}_k\}$ is all nonpositive or all nonnegative, then surely either \mathbf{n} or $-\mathbf{n}$ can not be written as a conical combination of $\{\mathbf{v}_k\}$, which means $\text{span}(\{\mathbf{v}_k\})$ can not be the full space. \square

One has consequently Algorithm 1 to test the STLC property of the vector fields $\{\dot{\mathbf{x}}_{\mathbf{Q}_k}\}$ at a given state \mathbf{x} .

Algorithm 1 Algorithm for STLC Testing

Input: State \mathbf{x} .
Output: True if \mathbf{x} is STLC; False if \mathbf{x} is not STLC.
1: Calculate all the vectors $\dot{\mathbf{x}}_{\mathbf{Q}_k}$, $k = 1, \dots, 24$;
2: **for** $i = 1, \dots, 23$ **do**
3: **for** $j = i + 1, \dots, 24$ **do**
4: **for** $k = 1, \dots, 24 \wedge k \neq i, j$ **do**
5: $c_k = \dot{\mathbf{x}}_{\mathbf{Q}_i} \times \dot{\mathbf{x}}_{\mathbf{Q}_j} \cdot \dot{\mathbf{x}}_{\mathbf{Q}_k}$;
6: **end for**
7: **if** $(c_1, \dots, c_{24} \geq 0) \vee (c_1, \dots, c_{24} \leq 0)$
8: $d_{ij} = \text{False}$;
9: **else**
10: $d_{ij} = \text{True}$;
11: **end if**
12: **end for**
13: **end for**
14: **Return** $\bigwedge_{i,j=1}^{24} d_{ij}$.

Relaxation Matrix Tomography on Chloroform

The basic liquid NMR relaxation theory can be found in [10]. In the rotating frame, it is routine to make a secular approximation by which the relaxation matrix would take a kite-like appearance. The underlying principle is that, the system energy level differences are much larger than the relaxation rates, so in the interaction picture, the cross relaxation parameters between the population subspace and the coherence subspace are added with fast oscillating phases. This effectively decoupled the population subspace relaxation from the coherence subspace relaxation. Secular approximation dramatically simplified the task of experimentally estimating the relaxation rates.

To be concrete, the relaxation dynamics can be decomposed as a direct sum of subspace dynamics (featured by the order of coherences)

- population subspace:

$$\frac{d}{dt} \begin{pmatrix} 1/4 \\ \langle ZI \rangle \\ \langle IZ \rangle \\ \langle ZZ \rangle \end{pmatrix} = \begin{bmatrix} 0 & 0 & 0 & 0 \\ -(4r_1 + 16r_4)\varepsilon & r_1 & r_4 & r_5 \\ -(4r_4 + 16r_2)\varepsilon & r_4 & r_2 & r_6 \\ -(4r_5 + 16r_6)\varepsilon & r_5 & r_6 & r_3 \end{bmatrix} \begin{pmatrix} 1/4 \\ \langle ZI \rangle \\ \langle IZ \rangle \\ \langle ZZ \rangle \end{pmatrix}, \quad (22)$$

- ^{13}C one-quantum coherence subspace:

$$\frac{d}{dt} \begin{pmatrix} \langle XI \rangle \\ \langle YI \rangle \\ \langle XZ \rangle \\ \langle YZ \rangle \end{pmatrix} = \begin{bmatrix} r_7 & 0 & r_9 & -\pi J \\ 0 & r_7 & \pi J & r_9 \\ r_9 & -\pi J & r_8 & 0 \\ \pi J & r_9 & 0 & r_8 \end{bmatrix} \begin{pmatrix} \langle XI \rangle \\ \langle YI \rangle \\ \langle XZ \rangle \\ \langle YZ \rangle \end{pmatrix}, \quad (23)$$

- H one-quantum coherence subspace:

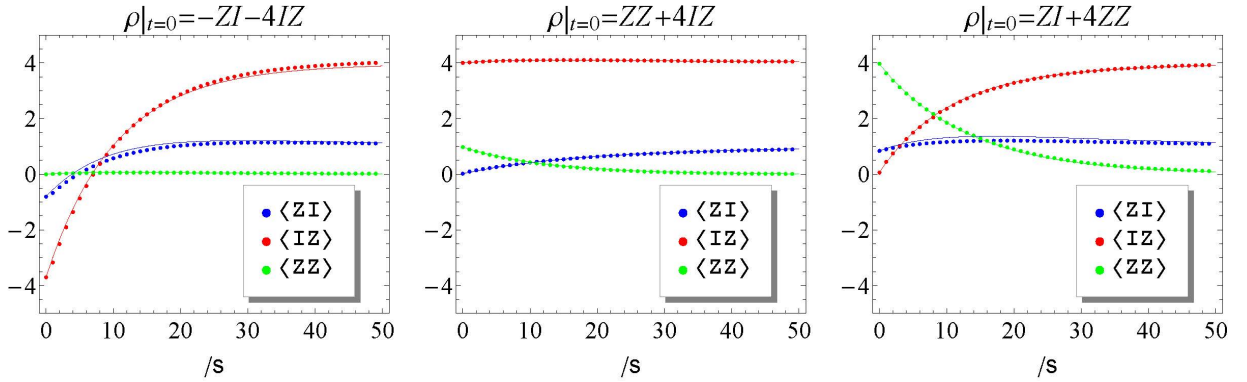
$$\frac{d}{dt} \begin{pmatrix} \langle IX \rangle \\ \langle IY \rangle \\ \langle ZX \rangle \\ \langle ZY \rangle \end{pmatrix} = \begin{bmatrix} r_{10} & 0 & r_{12} & -\pi J \\ 0 & r_{10} & \pi J & r_{12} \\ r_{12} & -\pi J & r_{11} & 0 \\ \pi J & r_{12} & 0 & r_{11} \end{bmatrix} \begin{pmatrix} \langle IX \rangle \\ \langle IY \rangle \\ \langle ZX \rangle \\ \langle ZY \rangle \end{pmatrix}, \quad (24)$$

- Subspace of zero- and double- quantum coherences:

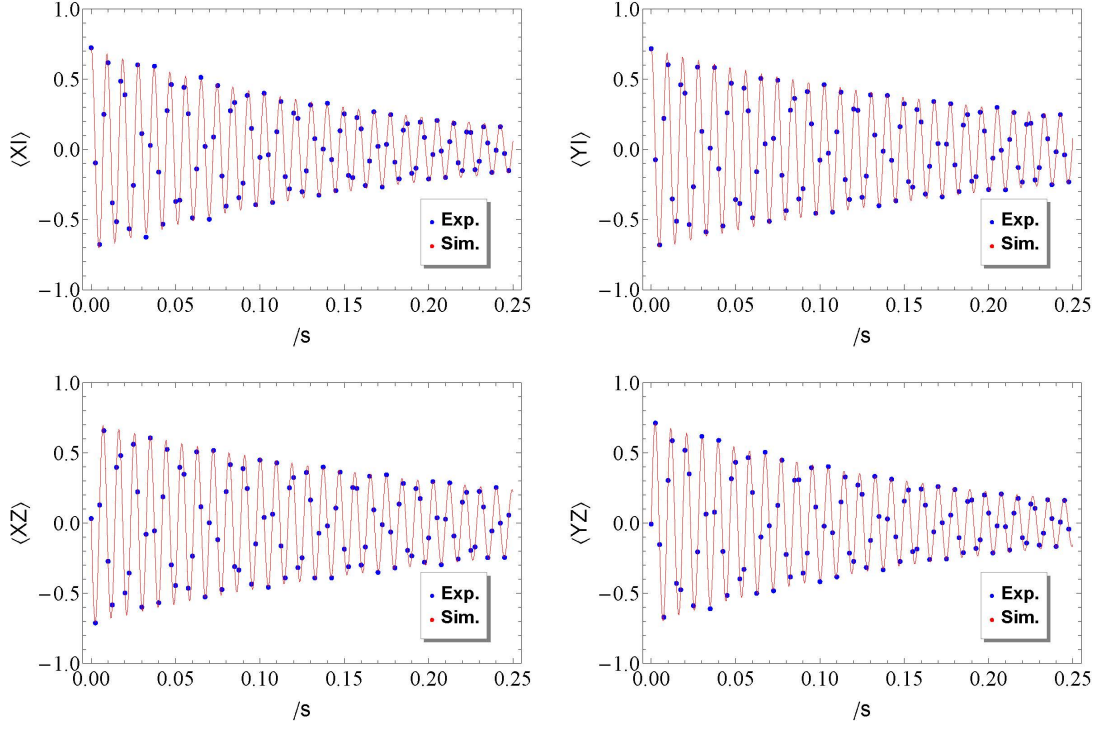
$$\frac{d}{dt} \begin{pmatrix} \langle XY \rangle \\ \langle YX \rangle \\ \langle XX \rangle \\ \langle YY \rangle \end{pmatrix} = \begin{bmatrix} r_{13} & -r_{14} & 0 & 0 \\ -r_{14} & r_{13} & 0 & 0 \\ 0 & 0 & r_{13} & r_{14} \\ 0 & 0 & r_{14} & r_{13} \end{bmatrix} \begin{pmatrix} \langle XY \rangle \\ \langle YX \rangle \\ \langle XX \rangle \\ \langle YY \rangle \end{pmatrix}, \quad (25)$$

where $\{r_k\}_{k=1,\dots,14}$ are relaxation rates including auto-relaxation rates and cross-relaxation rates. To estimate the relaxation rates, we first sample the system evolution trajectory (starting from a known initial state $\rho(0)$), then find values of the relaxation rates so that the simulated dynamics can match the observed trajectory. The fitting results are listed below (we have set $\epsilon = 1$)

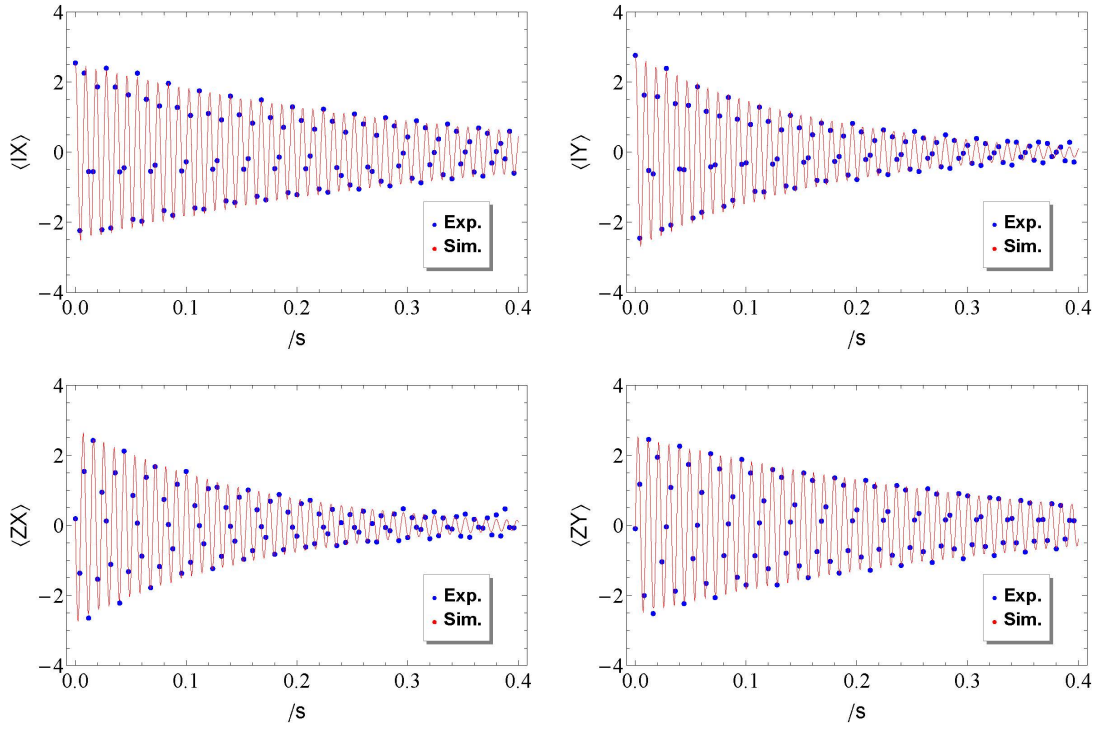
- $\{r_1, r_2, r_3, r_4, r_5, r_6\} \approx \{0.0532, 0.0918, 0.0798, 0.0212, 0.0000, 0.0022\}$



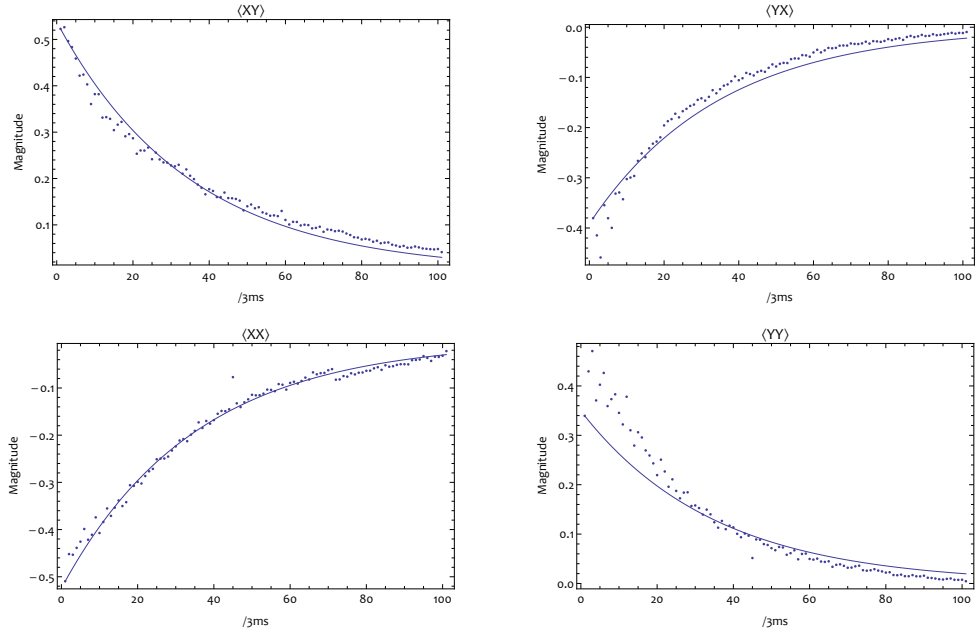
- $\{r_7, r_8, r_9\} \approx \{3.495, 6.536, 0.0100\}$



- $\{r_{10}, r_{11}, r_{12}\} \approx \{2.955, 6.118, 0.030\}$



- $\{r_{13}, r_{14}\} \approx \{9.523, 0.008\}$



Robustness of Periodic Control Method for PPS Preparation

Figure 2 shows the relative error of the prepared PPS due to imperfections of control fields present in the ^{13}C channel ($\delta_C = |B_{real}^C - B_{ideal}^C| / |B_{ideal}^C|$) and ^1H channel ($\delta_H = |B_{real}^H - B_{ideal}^H| / |B_{ideal}^H|$). The relative error is characterized by

$$\delta = \|\rho_{real} - \rho_{pps}\| / \|\rho_{pps}\|. \quad (26)$$

It can be easily seen that the periodic control method is quite robust to the control imperfections.

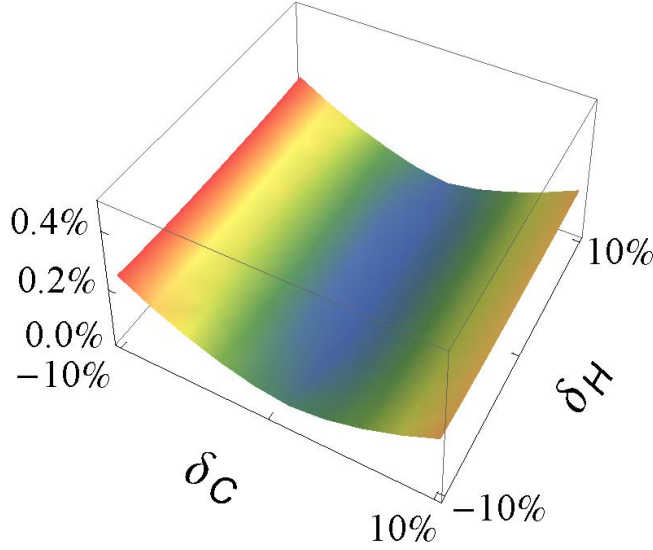


FIG. 3: Simulation result: relative error of the prepared PPS due to imperfections of control fields present in the ^{13}C channel and ^1H channel.

* Electronic address: xhpeng@ustc.edu.cn

† Electronic address: djf@ustc.edu.cn

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